

Electromagnetically induced coherent backscattering

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We demonstrate a strong coherent backward wave oscillation using forward propagating fields only. This is achieved by applying laser fields to an ultra-dispersive medium with proper chosen detunings to excite a molecular vibrational coherence that corresponds to a backward propagating wave. The physics then has much in common with propagation of ultra-slow light. Applications to coherent scattering and remote sensing are discussed.

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Quantum coherence [1, 2] has been shown to result in many counter-intuitive phenomena. The scattering via a gradient force in gases [3], the forward Brillouin scattering in ultra-dispersive resonant media [4, 5], electromagnetically induced transparency [6, 7, 8, 9], slow light [10, 11, 12, 13], Doppler broadening elimination [14], light induced chirality in nonchiral medium [15], a new class of entanglement amplifier [16] based on correlated spontaneous emission lasers [17, 18] and the coherent Raman scattering enhancement via maximal coherence in atoms [19] and biomolecules [20, 21] are a few examples that demonstrate the importance of quantum coherence.

In this Letter, we predict strong coherent backward scattering via excitation of quantum coherence between atomic or molecular levels. The developed approach can also be used to control the direction of the signal generated in coherent Raman scattering and other four-wave mixing (FWM) schemes.

Let us consider the four-wave mixing in a 3-level atomic medium. The pump and Stokes fields \mathcal{E}_1 and \mathcal{E}_2 (whose Rabi frequencies are defined as $\Omega_1 = \varphi_1 \mathcal{E}_1 / \hbar$ and $\Omega_2 = \varphi_2 \mathcal{E}_2 / \hbar$, where φ_1 and φ_2 are the dipole moments of the corresponding transitions) with wave vectors k_1 and k_2 and angular frequencies ν_1 and ν_2 induce a coherence grating in the medium (see Fig. 1) given by [2]

$$\rho_{cb} \sim -\Omega_1 \Omega_2^* \quad (1)$$

Let us stress that the ρ_{cb} coherence grating has an $\exp[i(k_1 - k_2)z]$ spatial dependence. In an ultra-dispersive medium (see Fig. 2) where fields propagate with a slow group velocity, the two co-propagating fields have wavevectors given by

$$k_1 \simeq k_1(\omega_{ab}) + \frac{\partial k_1}{\partial \nu_1}(\nu_1 - \omega_{ab}) = \omega_{ab}/c + (\nu_1 - \omega_{ab})/V_g, \quad (2)$$

where V_g is the group velocity of the first wave, ω_{ab} is the frequency of transition between levels a and b , and $k_2 = \nu_2/c$. Thus these two fields create a coherence grating

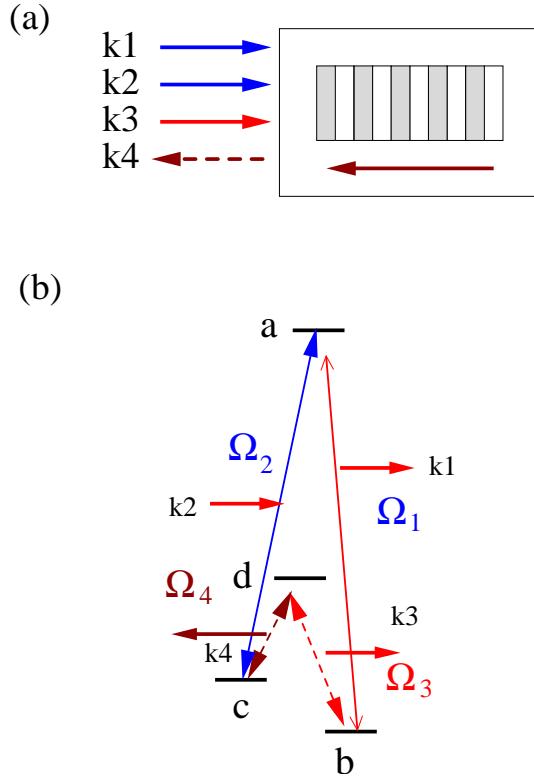


FIG. 1: (a) Co-propagating fields 1 and 2 induce coherent grating inside the medium. The field 3 propagating in the same direction will be scattered in the opposite direction because the coherence excited by fields 1 and 2 is propagating in the opposite direction (see Fig. 2). Level scheme, double- Λ (b), for implementation of coherent back scattering.

in the medium with spatial phase determined by $k_1 - k_2 = \omega_{cb}/c + (\nu_1 - \omega_{ab})/V_g$ which depends strongly on the detuning $\delta = \nu_1 - \omega_{ab}$. By properly choosing the detuning, δ , one can make $k_1 - k_2$ negative.

After the coherence ρ_{bc} is induced in the medium, a probe field \mathcal{E}_3 , with Rabi frequency $\Omega_3 = \varphi_3 \mathcal{E}_3 / \hbar$ and

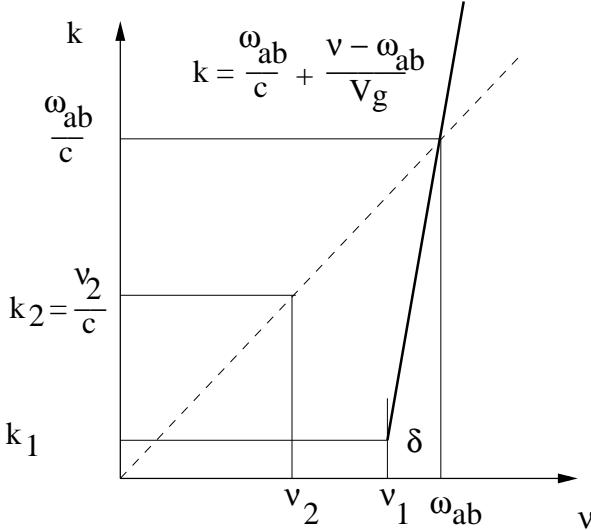


FIG. 2: Dispersion $k(\nu)$ of ultra-dispersive medium. Choosing $\delta = \nu_1 - \omega_{ab} = -V_g \omega_{cb}/c$, we can have $k_1 - k_2 < 0$ even if $\nu_1 > \nu_2$, thus the third field can be scattered opposite to the direction of propagation of the first two fields.

wave vector k_3 , scatters off that coherence to produce the signal field, Ω_4 . The signal field depends on the coherence and the input fields as

$$\frac{\partial}{\partial z} \Omega_4 \sim \rho_{cb} \Omega_3 \sim \Omega_1 \Omega_2^* \Omega_3 \sim e^{i(k_1 - k_2 + k_3 - k_4)z} \quad (3)$$

That is, the propagation direction of Ω_4 depends on the spatial phase of the ρ_{bc} coherence through the phase-matching condition $k_4 = k_1 - k_2 + k_3$ [22] while its frequency is determined by $\nu_4 = \nu_1 - \nu_2 + \nu_3$.

We here show that for dispersive media one can obtain a strong signal in the backward direction even when all three input fields propagate forward. This is contrary to the usual non-dispersive media results, where the phase-matching in the backward direction cannot be achieved for \mathcal{E}_1 , \mathcal{E}_2 , and \mathcal{E}_3 counter-propagating with respect to \mathcal{E}_4 [22].

To demonstrate this result, we write the interaction Hamiltonian of the system as

$$V_I = -\hbar [\Omega_2 e^{-i\omega_{act}} |a\rangle \langle c| + \Omega_1 e^{-i\omega_{ab}t} |a\rangle \langle b| + h.c.] \quad (4)$$

$$-\hbar [\Omega_3 e^{-i\omega_{db}t} |d\rangle \langle b| + \Omega_4 e^{-i\omega_{dc}t} |d\rangle \langle c| + h.c.] \quad (5)$$

where $\Omega_4 = \varphi_4 \mathcal{E}_4 / \hbar$ is the Rabi frequency of the signal field and ω_{ab} , ω_{ac} , ω_{db} , ω_{dc} are the frequency differences between the corresponding atomic or molecular energy levels. The time-dependent density matrix equations are given by

$$\frac{\partial \rho}{\partial \tau} = -\frac{i}{\hbar} [V_I, \rho] - \frac{1}{2} (\Gamma \rho + \rho \Gamma), \quad (6)$$

where Γ is the relaxation matrix. A self-consistent system also includes the field propagation equations

$$\frac{\partial \Omega_1}{\partial z} = -i\eta_1 \rho_{ab}, \quad \frac{\partial \Omega_2}{\partial z} = -i\eta_2 \rho_{ac}, \quad (7)$$

$$\frac{\partial \Omega_3}{\partial z} = -i\eta_3 \rho_{db}, \quad \frac{\partial \Omega_4}{\partial z} = +i\eta_4 \rho_{dc}, \quad (8)$$

where

$$\eta_j = \nu_j N \varphi_j / (2\epsilon_0 c) \quad (9)$$

are the coupling constants ($j = 1, 2, 3, 4$), N is the particle density of the medium, ϵ_0 the permittivity in vacuum.

The equations of motion for the density matrix elements of the polarization ρ_{ab} and the coherence ρ_{cb} are given by

$$\dot{\rho}_{ab} = -\Gamma_{ab} \rho_{ab} + i\Omega_1 (\rho_{aa} - \rho_{bb}) - i\rho_{cb} \Omega_2^*, \quad (10)$$

$$\dot{\rho}_{cb} = -\Gamma_{cb} \rho_{cb} + i\rho_{ca} \Omega_1 - i\rho_{ab} \Omega_2. \quad (11)$$

where $\Gamma_{ab} = \gamma_{ab} + i(\omega_{ab} - \nu_1)$; $\Gamma_{ca} = \gamma_{ca} - i(\omega_{ac} - \nu_2)$; $\Gamma_{cb} = \gamma_{cb} + i(\omega_{cb} - \nu_1 + \nu_2)$; ω_{cb} is the frequency of $c - b$ transition, and $\gamma_{\alpha\beta}$ are the relaxation rates at the corresponding atomic transitions. In the steady state regime, and assuming that $|\Omega_2| \gg |\Omega_1|$, almost all of the population remains in the ground level $|b\rangle$, $\rho_{bb} \simeq 1$. Let us consider the fields as plane waves: $\Omega_1(z, t) = \tilde{\Omega}_1(z, t) \exp(ik_1 z)$, $\Omega_2(z, t) = \tilde{\Omega}_2(z, t) \exp(ik_2 z)$, where $\tilde{\Omega}_1(z, t)$ and $\tilde{\Omega}_2(z, t)$ are the slowly varying envelopes of the fields Ω_1 and Ω_2 in space, while $k_1 = \nu_1 [1 + \chi_{ab}(\nu_1)]/c$ and $k_2 = \nu_2 [1 + \chi_{ac}(\nu_2)]/c$. The susceptibilities are $\chi_{ab} = \eta_1 \rho_{ab} / \Omega_1 = (\nu_1 - \omega_{ab}) / 2\pi V_g$ and $\chi_{ac} = \eta_2 \rho_{ac} / \Omega_2 \simeq 0$. By solving the self-consistent system of Maxwell's equations (7,8) and the density matrix equations (10,11), we obtain Eq. (2) for the wavevectors, where $V_g \simeq c/\eta_1 |\Omega_2|^2$ is the group velocity of the optical field Ω_1 . Thus, the spatial dependence of ρ_{cb} is determined by

$$\Delta k = k_1 - k_2 = \frac{\nu_1 - \nu_2}{c} + \frac{\nu_1 - \omega_{ab}}{V_g}. \quad (12)$$

The signal field Ω_4 is generated by the polarization ρ_{db} of the transition it couples (Eq. 8). The equation of motion for this polarization element reads

$$\dot{\rho}_{db} = -\Gamma_{db} \rho_{db} + i\Omega_4 (\rho_{dd} - \rho_{bb}) - i\rho_{cb} \Omega_3^*, \quad (13)$$

where $\Gamma_{db} = \gamma_{db} + i(\omega_{db} - \nu_4)$, and ν_4 is the frequency of generated field. In the steady-state regime and for $|\Omega_4| \ll |\Omega_3|$, the field Ω_4 at the output of the cell is given by

$$\Omega_4 = \int_0^L dz e^{i(k_4 - \Delta k - k_3)z} \frac{\eta_4 \rho_{cb} \tilde{\Omega}_3^*}{\Gamma_{db}} \quad (14)$$

where L is the length of the cell. Note here that Eq.(14) is valid if the field $|\Omega_3|$ does not change coherence ρ_{cb} via power broadening which is true if $|\Omega_3|^2 \ll |\Omega_1|^2 + |\Omega_2|^2$.

Hence, after integrating Eq.(14), we obtain for the scattered field Ω_4

$$\Omega_4 = \left[\frac{\sin(k_4 - k_3 - \Delta k)L}{(k_4 - k_3 - \Delta k)L} \right] \frac{\eta_4 L \Omega_3}{\Gamma_{db}} \quad (15)$$

where the expression in the brackets describes the phase-matching and determines the direction in which the signal field is generated.

The most interesting effect following from Eq.(15) is the coherent backscattering. Indeed, even when all three input fields propagate forward, one may observe a backscattered signal field by satisfying the condition

$$k_3 + \Delta k = k_3 + \frac{\nu_1 - \omega_{ab}}{V_g} < 0, \quad (16)$$

and for appropriate detuning, $\nu_1 - \omega_{ab} < 0$, this inequality can be met. That is, in order to obtain phase-matching in the backward direction, we have to satisfy

$$k_3 + \Delta k < -k_4, \quad (17)$$

which can be rewritten as

$$-\frac{\nu_1 - \omega_{ab}}{V_g} > k_3 + k_4. \quad (18)$$

Hence, in order to demonstrate the effect, the detuning δ should meet the condition

$$\delta = -(k_3 + k_4)V_g \simeq -2k_4V_g. \quad (19)$$

It is useful to rewrite the condition in terms of susceptibility for the probe field, indeed,

$$k_1 = \frac{\nu_1}{c}n_1 \simeq \frac{\nu_1}{c}(1 + c\frac{\nu_1 - \omega_{ab}}{\nu_1 V_g}) = \frac{\nu_1}{c}(1 - c\frac{2k_4}{\nu_1}), \quad (20)$$

then,

$$\chi_{ab} = 2(n_1 - 1) = -4\frac{\lambda_{ab}}{\lambda_{db}}, \quad (21)$$

for gases $\chi_{ab} \ll 1$, so $\lambda_{ab} \ll \lambda_{db}$, i.e. the effect can be implemented for scattering of IR fields. Then, for the Doppler broadened EIT media as shown in [23, 24], we can write

$$\chi_{ab}(\delta) \simeq \frac{3\lambda_{ab}^3 N}{8\pi^2} \left(\frac{\gamma_r \delta}{|\Omega_2|^2} + i \frac{\gamma_r \Delta_D \delta^2}{|\Omega_2|^4} \right), \quad (22)$$

where Δ_D is the Doppler width; γ_r is the radiative decay rate. Thus, for detuning smaller than the EIT width $|\delta| \leq |\Omega_2|^2 / \sqrt{\gamma_r \Delta_D}$, absorption can be neglected, and

$$\frac{3\lambda_{ab}^2 N \gamma_r \delta}{16\pi |\Omega|^2} = -2k_4, \quad (23)$$

then, the atomic or molecular density is given by

$$N = \frac{32\pi k_4 |\Omega_2|^2}{3\lambda_{ab}^2 \gamma_r |\delta|} \simeq \frac{32\pi k_4}{3\lambda_{ab}^2} \sqrt{\frac{\Delta_D}{\gamma_r}}. \quad (24)$$

There are several schemes to demonstrate the effect. For example, the double-Lambda scheme can be implemented in molecular rotational levels (see Fig. 1a). Moreover, the effect can be implemented in the ladder- Λ using

molecular vibrational levels (see Fig. 3a). The phase-matching condition should be slightly modified for the scheme as $k_4 = k_1 - k_2 - k_3$. Also, the phenomenon can be demonstrated in a V- Λ scheme that can be realized in atomic levels (see Fig. 3b, for Rb atoms, $b = 5S_{1/2}$, $c = 7D_{3/2,5/2}$, $a = 5P_{1/2,3/2}$, $d = 8P_{1/2,3/2}$), and phase-matching condition has a form $k_4 = k_1 + k_2 - k_3$. Let us note that the requirement for detuning in all cases is $\delta/V_g = -2k_4$ and the use of Eq. (24) to estimate molecular or atomic density is still valid.

Examples of systems to this effect could be seen we mention molecules NO (a resonant transition at 236 nm, $A^2\Sigma^+ - X^2\Pi$), NO_2 (a resonant transition at wavelength 337 nm), and atomic Rb vapor (EIT and CPT have been recently demonstrated for molecules, see [26]). The required molecular density of NO and NO_2 molecules is $N \simeq 1.2 \cdot 10^{13} \text{ cm}^{-3}$ if one can use transition between rotational levels $\simeq 10 \text{ cm}^{-1}$. Using vibrational IR transitions for NO (vibration frequency of 1900 cm^{-1}) at $5.26 \mu\text{m}$ and for NO_2 (vibrational frequency of 750 cm^{-1}) $13.3 \mu\text{m}$, the densities are $N = 8 \cdot 10^{15} \text{ cm}^{-3}$ and $N = 1.4 \cdot 10^{15} \text{ cm}^{-3}$, correspondingly. For atomic Rb vapor, wavelengths are $\lambda_1 = 780 \text{ nm}$, $\lambda_2 = 565 \text{ nm}$, $\lambda_3 = 335 \text{ nm}$, $\lambda_4 = 23.4 \mu\text{m}$, and the atomic density is $N = 1.4 \cdot 10^{13} \text{ cm}^{-3}$.

The intensity needed for EIT is determined by condition $|\Omega|^2 \gg \gamma_{bc}\Delta_D$ which corresponds to laser intensity of the order of 1 mW/cm^2 for atoms and of the order of 10 W/cm^2 for molecules because the dipole moment is two orders of magnitude smaller for molecules. These conditions are realistic and well-suited for an experimental implementation.

Several applications of the effect can be envisioned, like in nonlinear CARS microscopy [27], while the controlling of coherent backscattering could provide a new tool for creating an image. A variation in the molecular density would modify the intensity of the signal in both the forward and the backward direction. Additionally, Eq.(15) also allows one to control the direction of the generated signal field and thus provide an all-optical control when scanning an optical field over an object.

In conclusion, we theoretically predict strong coherent scattering in the backward direction while using only forward propagating fields. This is achieved by exciting atomic or molecular coherence by properly detuned fields, in such a way that the resulting coherence has a spatial phase corresponding to a backward, counter-propagating wave. Applications of the technique to coherent scattering and remote sensing are discussed. The method holds promise for observation induced scattering in a backward direction with application to CARS microscopy.

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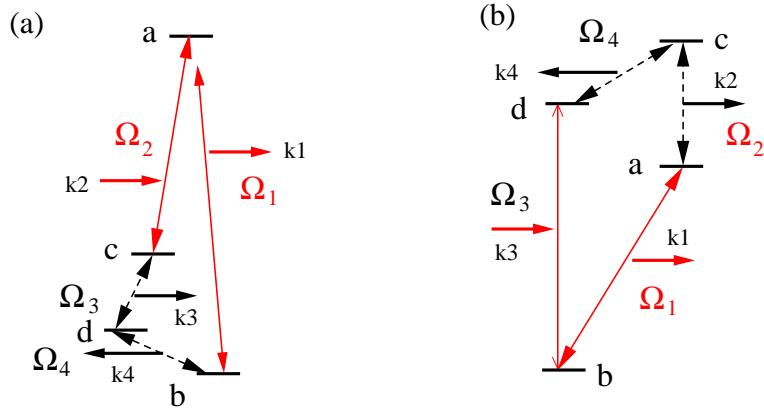


FIG. 3: Implementation: molecular systems, (a) vibrational levels; co-propagating fields 1 and 2 induce coherent between vibrational levels. The field 3 propagating in the same direction will be scattered in the opposite direction. (b) Atomic Rb scheme for implementation of coherent back scattering.

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